

Tracer Dispersion in Porous Media with Spatial Correlations

Hernán A. Makse ¹⁻³, José S. Andrade Jr. ¹, and H. Eugene Stanley ³

¹Departamento de Física, Universidade Federal do Ceará, 60451-970 Fortaleza, Ceará, Brazil

²Schlumberger-Doll Research, Old Quarry Road, Ridgefield, CT 06877

³Center for Polymer Studies and Physics Dept., Boston University, Boston, MA 02215

(February 7, 2008)

We analyze the transport properties of a neutral tracer in a carrier fluid flowing through percolation-like porous media with spatial correlations. We model convection in the mass transport process using the velocity field obtained by the numerical solution of the Navier-Stokes and continuity equations in the pore space. We show that the resulting statistical properties of the tracer can be approximated by a Lévy walk model, which is a consequence of the broad distribution of velocities plus the existence of spatial correlations in the porous medium.

The phenomenon of hydrodynamic dispersion—the unsteady transport of a neutral tracer in a carrier fluid flowing through a porous material—has been widely investigated in the fields of petroleum and chemical engineering [1,2]. One can identify different regimes of tracer dispersion according to the Péclet number $Pe \equiv v\ell/D_m$, which is the ratio between the typical time for diffusion ℓ^2/D_m and the typical time for convection ℓ/v . Here v is the velocity of the carrier fluid, ℓ a characteristic length scale of the porous media, and D_m the molecular diffusivity of the tracer.

In the small Péclet number regime, molecular diffusion dominates the way in which the tracer samples the flow field. In the large Péclet number regime, also called *mechanical dispersion*, convection effects are significant; the tracer velocity is approximately equal to the carrier fluid velocity, and molecular diffusion plays little role. The tracer samples the disordered medium by following the velocity streamlines. In a random walk picture, we may think of a tracer particle following the direction of the velocity field, and taking steps of length ℓ and duration ℓ/v .

The classical approach to model dispersion in porous media is to consider microscopically disordered and macroscopic isotropic and homogeneous porous materials. Under these conditions, dispersion is said to be *Gaussian* and the phenomenon can be mathematically represented in terms of the convection-diffusion equation [2]. This traditional formalism, which is valid for Euclidean geometries, cannot be adopted to describe the global behavior of hydrodynamic dispersion in heterogeneous systems. Specifically, in the case of percolation porous media, the breakdown of the macroscopic convective-diffusion description is a direct consequence of the self-similar characteristic of the void space geometry.

Here we discuss the interesting physics that arises when the tracer moves in a flow field with a very broad velocity distribution. Consider, e.g., fluid flow in percolation clusters near the percolation threshold—a model system relevant to a porous medium with stagnant small-velocity zones that are linked with large-velocity zones. In this case the typical time for convection ℓ/v is without bound

since the velocity can be arbitrarily small in some fluid elements of the void space. Saffman showed [1] that the mean square duration of a tracer step is not finite but diverges logarithmically unless an upper cut-off is introduced into the typical time step. This upper cut-off is imposed by the mass transport mechanism of molecular diffusion.

Molecular diffusion is expected to affect the tracer motion in two ways [1]:

(i) A quantity of material may cross from one streamline with fluid velocity v to another by *lateral* diffusion if the time step for convection ℓ_{\parallel}/v is larger than t_1 , where $t_1 = \ell_{\perp}^2/2D_m$ is the characteristic time for molecular diffusivity effects to become appreciable [3] and ℓ_{\parallel} and ℓ_{\perp} are the longitudinal and lateral pore lengths, respectively (with respect to the flow direction) [1]. Thus, if $\ell_{\parallel}/v \gg t_1$, the tracer has enough time to diffuse across the pore, and the time step associated with such a move is $\Delta t = t_1$. When $\ell_{\parallel}/v \ll t_1$, the time duration of a convective step is smaller than the time required for molecular diffusion, and the tracer moves with the carrier fluid taking a step of duration $\Delta t = \ell_{\parallel}/v$.

(ii) An amount of material may be transported by diffusion *along the pore*. The same considerations as in point (i) lead to a time step $\Delta t = \ell_{\parallel}/v$ in which convection dominates when $\ell_{\parallel}/v \ll t_0 = \ell_{\parallel}^2/2D_m$. Here the typical length scale is the longitudinal length of the pore ℓ_{\parallel} . If $\ell_{\parallel}/v \gg t_0$, diffusion dominates and the tracer takes a time step $\Delta t = t_0$.

Here we propose a model of tracer dispersion in a porous medium. The porous medium is composed of blocks of impermeable material that occupy, with a given probability p , a square lattice. We consider a lattice at the site percolation threshold, so an incipient spanning cluster is formed that connects the two ends of the lattice. Previous studies modeled the convective local “bias” for the movement of the neutral tracer in the porous media assuming Stokes flow [2]. Even at macroscopically small Reynolds conditions, this assumption might be violated in real flow through porous media, specially in the case of heterogeneous materials (e.g., percolation-like structures) where a broad distribution of pore sizes can lead to a broad distribution of local fluxes.

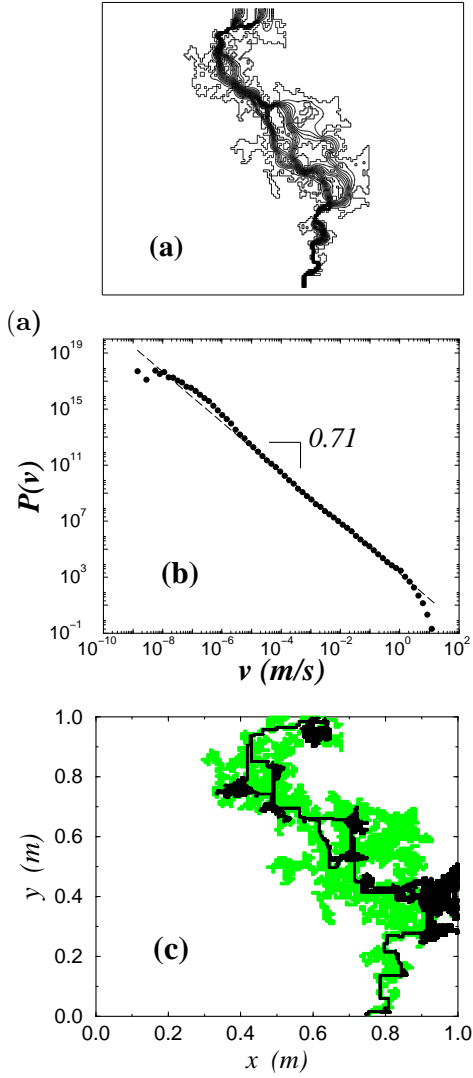


FIG. 1. (a) Typical stream-lines of the velocity field in a correlated percolation cluster. (b) Velocity magnitudes probability distribution averaged over 5 realizations of the percolation clusters. (c) Tracer diffusion in the porous medium shown in (a), for $Pe = 1.7$. We release five walkers and the black dots indicate the sites visited at least one time by the walkers.

As a consequence, inertial effects might be locally relevant. To avoid this problem, we use the steady-state velocity field obtained by solving the full set of Navier-Stokes in the percolation geometry. Then we study the transport properties of a dynamically-neutral tracer moving in the flow field.

We treat the competition between the effects of convection and diffusion. The velocity field presents a broad scale-invariant power-law distribution of magnitude values, and we find that there are regions of very small velocity in which the tracer can be trapped. If convection is important, the tracer follows the stream-lines of the fluid. When a very small velocity region is reached, molecular diffusion effects cannot be neglected, since by diffusion the tracer may access the stagnant zones—where it then

spends a long time. We shall see that due to the existence of these stagnant zones, the statistical properties of the tracer—e.g., the first-passage time and the root mean square displacement—can be understood using a Lévy walk model for the tracer motion. The existence of Lévy statistics is also related to the geometrical properties of the medium—whether it is correlated or uncorrelated in the occupancy variables of the percolation cluster.

We start by describing the disordered medium and the velocity field. Our basic model of a porous medium is a percolation model [4] modified to introduce correlations among the occupancy units [5]. We assume the existence of correlations because we obtain a better mathematical representation of transport properties—such as sandstone permeability—by assuming the presence of long-range correlations in the permeability fluctuations of the porous rock [6]. The permeability of rocks such as sandstone can fluctuate over short distances, and these fluctuations significantly affect any fluid flow through the rock. Previous models assumed that these fluctuations were random and without short-range correlations. However, permeability is not the result of a simple random process. Geologic processes, such as sand deposition by moving water or wind, impose their own kind of correlations.

The mathematical approach we apply to describe this situation is correlated percolation. In the limit where correlations are so small as to be negligible [4], a site at position \vec{r} is occupied if the occupancy variable $u(\vec{r})$ is smaller than the occupation probability $0 \leq p \leq 1$; the variables $u(\vec{r})$ are uncorrelated random numbers with uniform distribution in the interval $[0, 1]$. To introduce long-range power-law correlations among the variables, we convolute the uncorrelated variables $u(\vec{r})$ with a suitable power law kernel [7], and define a new set of occupancy variables $\eta(\vec{r})$ with long-range power-law correlations that decay as $r^{-\gamma}$, where $r \equiv |\vec{r}|$ (in the following we will set $\gamma = 0.4$).

We solve the full set of Navier Stokes and continuity equations at the percolation threshold of a square lattice with 64×64 cells and cell edge $L = 1$ m. Grid element lengths with $1/4$ of the solid cell edge, $\ell_{\parallel} = \ell_{\perp} = \ell = L/256$, have been adopted to discretize the governing balance equations within the pore space domain [8]. Figure 1a shows a typical velocity field, while Fig. 1b shows the probability distribution of the velocity magnitudes averaged over five realizations of the percolation clusters. We find that the data are well fit by a broad power-law of the type [8]

$$P(v) \sim v^{-0.71}. \quad (1)$$

Next we analyze the transport properties of a neutral tracer moving in the fluid. We use a discrete random walk model for the tracer motion. Following the Saffman theory of dispersion in porous media, we define the walker motion as a competition between flow-driven convection and molecular diffusion. To allow for comparison among

different regimes of tracer dispersion, we define a macroscopic Péclet number as $Pe \equiv v_{in}\ell/D_m$, where $v_{in} = 1$ m/s is the fluid velocity at the inlet boundary of the lattice. At a given position \vec{r} in the pore space, we define the time scale for convection $t_c \equiv \ell/v(\vec{r})$. We choose a convective or diffusive move, and a corresponding time step Δt according to:

$$t_c < t_d, \quad \text{convection, } \Delta t = t_c \quad (2)$$

$$t_c > t_d, \quad \text{diffusion, } \Delta t = t_d. \quad (3)$$

Here $t_d \equiv \ell^2/2D_m = Pe\ell/(2v_{in})$ is the characteristic time above which diffusion effects become relevant. If the convection move is accepted, then the tracer moves to the nearest-neighbor site in the direction given by the velocity of the fluid and the clock is updated according to $t \rightarrow t + t_c$. If the diffusion move is accepted, then the tracer moves to one of the four nearest-neighbor positions with equal probability and the clock is updated according to $t \rightarrow t + t_d$.

We next discuss the case of large Péclet number, $Pe = 1.7$, so the value of t_d is such that diffusion only occurs in regions of small fluid velocity. Typical tracer trajectories are shown in Fig. 1c. We see that the tracer particles perform walks with very long straight trajectories followed by periods where they get trapped in small velocity zones. These “stagnant zones” in the pore space differ significantly from the dangling ends of the analogous electrical problem (i.e., the parts of the infinite cluster connected by only one site to the backbone). The tracer enters these regions by diffusion, and requires a long time to escape. After escaping, the particle performs another almost ballistic trajectory until it penetrates into the next small velocity region. The tracer trajectory resembles a quasi-one-dimensional channel of “tubes and blobs.” The “tubes and blobs” picture is the analog for this problem of the traditional “links and blobs” picture associated with anomalous diffusion in percolation clusters [9,10].

We analyze the transit time, i.e., the average time required for the tracer to traverse a given distance x from the inlet line, $0 < x < L$, for different Péclet numbers. We find (Fig. 2a) that the transit times follow a power law

$$\langle t \rangle \sim x^\beta \quad (4)$$

where $\beta \simeq 1.26$ when $Pe = 1.7$.

In the “tubes and blobs” picture, we define a tube as the set of steps taken by the tracer following a fixed direction, and we analyze the statistical distribution of the tube length s . In stagnant zones where diffusion is dominant, the tracer is expected to change direction every time step, so that $s \simeq \ell$. In regions in which convection dominates, the tracer moves in ballistic trajectories limited only by impermeable obstacles. Since long-range correlated clusters are very compact (Fig. 1a), we expect $s \gg \ell$ and the tube length distribution to be broad. Both expectations are corroborated by our calculations.

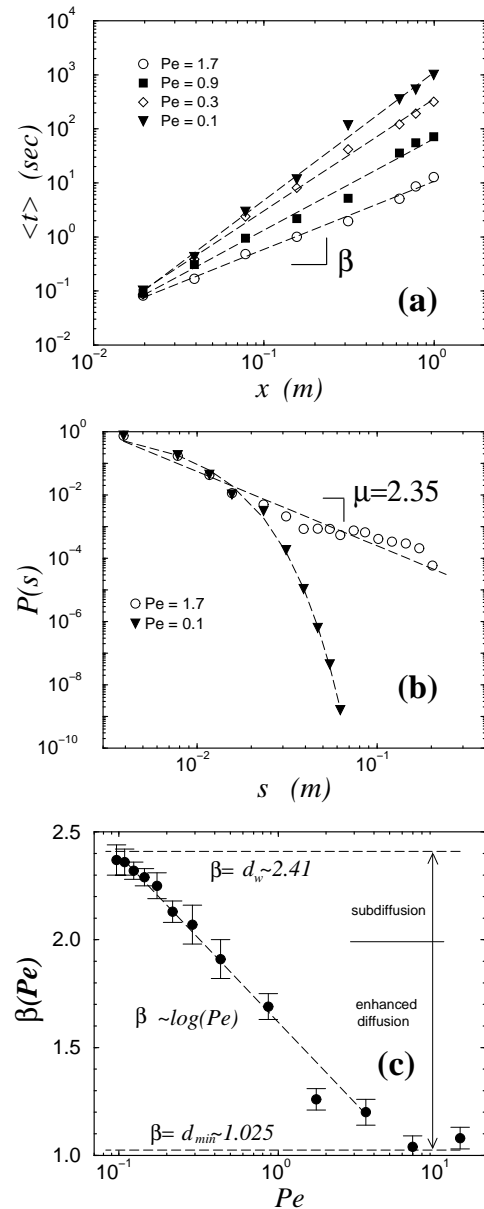


FIG. 2. (a) Transit times for different Péclet numbers, averaged over five realizations of the percolation clusters. (b) Probability distributions of steps for the case of $Pe = 1.7$ (power-law distribution) and $Pe = 0.1$ (Gaussian distribution). (c) Transit time exponent as a function of the Péclet number.

Figure 2b shows the probability distribution $P(s)$ for two different values of Pe . We find that for sufficiently large Pe , the step lengths follow a scale invariant power law distribution

$$P(s) \sim s^{-2.35} \quad [Pe = 1.7], \quad (5)$$

while for small Pe values, when diffusion is the dominant mechanism in the entire pore space, the distribution is Gaussian

$$P(s) \approx e^{-\frac{1}{2}(s/s_0)^2} \quad [Pe = 0.1], \quad (6)$$

with s_0 a characteristic jump length.

In case (5), the step lengths statistics can be considered as a Lévy walk, i.e., a random walk process in which the jump distribution is a power law [11]

$$P(s) \sim s^{-\mu}. \quad (7)$$

A random walker with a distribution (7) travels a typical distance $r \sim t^{2-\mu/2}$, when $2 < \mu < 3$. Thus, the transit time for a Lévy walker with jump statistics given by (7) is [11]

$$\langle t \rangle \sim x^{2/(4-\mu)}. \quad (8)$$

For $\mu = 2.35$ —the value we find in our simulations for $Pe = 1.7$ —we obtain $\langle t \rangle \sim x^{1.21}$, which agrees with the scaling found when we calculate the transit time directly, $\langle t \rangle \sim x^{1.26}$ from Fig. 2a, and confirms the validity of the Lévy walk picture as an accurate description of the tracer motion at large Pe .

The transit time exponent β is not universal and depends on Pe (Fig. 2c). In fact we find that the Lévy statistics approximates well the value of β in the entire enhanced diffusion regime $1 < \beta < 2$, while in the sub-diffusion regime $\beta > 2$, the Lévy statistics Eq. (8) ceases to be valid. Moreover, we expect two limiting regimes. If convection dominates completely (mechanical dispersion), then the tracer should follow the minimum path along the spanning percolation cluster. The minimum path length ℓ_{\min} scales as $\ell_{\min} \sim x^{d_{\min}}$ where d_{\min} is the fractal dimension of the minimum path distance between two points separated by a linear distance x [4]. If the tracer moves with a constant velocity, we can identify the minimum path distance with the transit time, so $\beta = d_{\min}$. This is the lower limit of the transit time exponent, and we confirm this prediction since we obtain $\beta \gtrsim d_{\min}$ when Pe is large (Fig. 2c) [12].

The other limit at larger diffusivities—the anomalous diffusion case [10]—corresponds to the regime dominated completely by diffusion, and the transit time scales as $\langle t \rangle \sim x^{d_w}$, where d_w is the random walk fractal dimension. The value d_w depends on the degree of correlation, with $d_w = 2.87$ for the uncorrelated percolation limit [4] and $d_w = 2.41$ [5] for the correlated percolation problem we study ($\gamma = 0.4$). We see that the limiting cases of our calculations agree with these predictions (Fig. 2c). Between these two limiting cases, we find that the transit time exponent can be approximated by

$$\beta(Pe) \sim \log(Pe). \quad (9)$$

We also perform simulations on uncorrelated percolation clusters. We find an enhanced diffusion regime and a sub-diffusion regime as well. However, due to the tortuosity of the uncorrelated percolation clusters at the threshold, the distribution of steps is not a scale-free power-law,

as we find in the case of enhanced diffusion in correlated clusters Eq. (5). Thus, we conclude that the Lévy statistics found in the case of dispersion in correlated clusters is a by-product of the dynamical properties of the tracer moving in a broadly distributed velocity field plus the geometrical properties of the particular porous medium treated here. The compact features of long-range correlated percolation clusters allows the tracer to perform large ballistic steps without encountering obstacles during the random walk process.

In summary, we find that, at sufficiently large Péclet numbers, there is a regime of dispersion for correlated porous media in which the trajectory of the tracer particle should be better described by a Lévy statistics Eq. (5) instead of the Gaussian behavior Eq. (6). Interestingly, this fact should be relevant to elucidate the mass and momentum transport mechanisms responsible for the dispersion regime called “holdup dispersion” [2]. Tracer experiments indicate that this regime of strong dependence between dispersion measurements and Péclet number is typical of percolation-like porous materials.

-
- [1] P. G. Saffman, *J. Fluid Mech.* **6**, 321 (1959).
 - [2] See, e.g., the comprehensive review M. Sahimi, *Flow and Transport in Porous Media and Fractured Rock* (VCH, Boston, 1995) and extensive references therein.
 - [3] G. I. Taylor, *Proc. R. Soc. London, Ser. A* **219**, 186 (1953); R. Aris, *Proc. R. Soc. London, Ser. A* **235**, 67 (1956).
 - [4] A. Bunde and S. Havlin, eds., *Fractals and Disordered Systems*, 2nd ed. (Springer, Berlin 1996).
 - [5] S. Prakash, S. Havlin, M. Schwartz, and H. E. Stanley, *Phys. Rev. A* **46**, R1724 (1992); H. A. Makse, *et al.*, *Physica A* **233**, 587 (1996).
 - [6] T. A. Hewett, SPE 15386, 61st Ann. Tech. Conf. of SPE, New Orleans, Louisiana (1986); M. Sahimi, *J. Phys. I (France)* **4**, 1263 (1994); H. A. Makse, *et al.*, *Phys. Rev. E* **54**, 3129 (1996).
 - [7] H. A. Makse, S. Havlin, M. Schwartz, and H. E. Stanley, *Phys. Rev. E* **53**, 5445 (1996).
 - [8] J. S. Andrade Jr., *et al.*, *Phys. Rev. Lett.* **79**, 3901 (1997).
 - [9] H. E. Stanley, *J. Phys. A* **10**, L211 (1977); A. Coniglio, *Phys. Rev. Lett.* **46**, 250 (1981).
 - [10] S. Havlin and D. Ben-Avraham, *Adv. Phys.* **36**, 695 (1987).
 - [11] M. F. Shlesinger, G. Zaslavsky, and U. Frisch, eds., *Lévy Flights and Related Topics in Physics* (Springer, Berlin, 1995); J. Klafter, A. Blumen, and M. F. Shlesinger, *Phys. Rev. A* **35**, 3081 (1987); J.-P. Bouchaud and A. Georges, *Phys. Rep.* **195**, 127 (1990).
 - [12] Notice that the value $d_{\min} \simeq 1.025$ corresponds to the correlated percolation value [5], 10% smaller than the value $d_{\min} \simeq 1.135$ for the uncorrelated percolation problem [4].